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MERCURY EMISSIONS FROM COAL-FIRED POWERPLANTS**ABSTRACT**

Mercury is a neurotoxic heavy metal that pose a risk to the public as well as the environmental health. It is a naturally occurring element that is released into the environment from the natural or anthropogenic emissions. Among the anthropogenic emissions, coal-fired powerplants account for approximately 50 percent of the emissions because of the lack of regulations for the emissions from these facilities. The other larger sources of mercury emissions such as municipal waste combustors and medical waste incinerators are subjected to the stringent regulations thereby minimizing their total contribution. Mercury is a natural component of coal and is released into the environment during the combustion of coal. Use of coals that contain less mercury can reduce the total mercury emissions. Mercury is liberated into the environment in several forms that can be categorized into elemental, inorganic and organic mercury. All these forms differ in their degree of toxicity and the utmost toxic species is the organic mercury. After emitting from the powerplants, mercury circulates in the atmosphere and gets deposited on the land and surface waters where the toxic species of mercury are formed. Reducing the mercury emissions from the powerplants can reduce the risk of this neurotoxic metal on humans. The use of control technology can significantly reduce the total mercury emissions in the future in a timely and cost effective manner.

1. INTRODUCTION

Mercury is a heavy metal that is hazardous to the humans and it is ranked third on Agency for Toxic Substances and Disease Registry's priority list after arsenic and lead (ATSDR, 2015). Mercury is released into the environment through natural processes or human activities. The natural sources of mercury emissions are from the volcanic eruptions and from the ocean emissions. Anthropogenic sources includes the emissions from the fuels or from the industrial processes. Among the human activities, more than 50% of mercury is released from coal-fired power plants (USEPA, 2015).

According to the USEPA (2015), in 1990, two-thirds of the mercury emissions in the United States is from three sectors that include medical waste incinerators, municipal waste combustors and power plants. In 2005, the emissions from medical waste incinerators and municipal waste combustors is reduced by more than 95 percent but in contrast, the mercury emissions from coal fired power plants is reduced only by 10 percent. Unlike other sources of mercury emissions, coal fired powerplants has no limitations for mercury emissions that causes a significant increase in the emissions (USEIA, 2015). Because the mercury emissions are concentrated locally, the coal fired powerplants are the major source of local impacts of mercury (Driscoll, 2007). There are certain species of mercury that travel for longer distances and result in global contaminations (Kuiken & Stadler, 2003).

2. MERCURY IN COAL

Mercury occurs naturally in coal. The average concentrations of mercury in the coals range from 0.12 to 0.28 $\mu\text{g/g}$ (NJ DEPE, 1993). Mercury is released from coal during the combustion process. The average concentration of mercury that is present in the gas emissions from the power plant is less than 10 $\mu\text{g/m}^3$ (Prestbo & Bloom, 1995). Mercury content varies based on the coal types. According to Yudovich & Ketris, (2005), there are more than three forms of mercury in coals that include clays, organic matter and sulfides. There are also many components in which mercury resides which include biogenic, sorption, terrigenous, clastics and diagenic or epigenic mineral fractions based on the indications of use of coal. According to Toole-O'Neil, Tewalt, Finkelman, & Akers, (1999), in-ground coal has higher concentration of mercury and the cleaned and shipped coal has a lower concentration which is due to the process of coal cleaning that reduces the concentration of mercury by approximately 37 percent. The conventional coal cleaning process reduced 47 percent of mercury from the coal whereas advanced cleaning by the use of advanced floatation or specific gravity separation reduced about 84 percent of mercury (Pavlish et al., 2003). The remaining mercury is liberated during coal combustion during the production of energy. The mercury that is present after coal cleaning is called as authigenic mercury and it includes organic matter and sulfides which determines the amount of mercury distributed in coal (Yudovich & Ketris, 2005). As the concentration of mercury is reduced, the coal will become less efficient for thermal productivity (Pavlish et al., 2003).

The amount of mercury present in pyritic forms of coal that was studied indirectly by correlation between sulfur and mercury concentrations, or directly by the sulfide analyses concluded that there is strong association of mercury content with the pyritic coal (Huggins & Huffman, 1996). In the coals with higher concentrations of mercury, there is an increase in the amounts of mercuric sulfides and mercury bearing zinc and lead sulfides (Yudovich & Ketris, 2005). Mercury can occur in pyritic coals due to a number of processes such as, the presence of small inclusions of cinnabar in pyrites. During the formation of pyrite in hydrothermal solutions, mercury can be introduced into it as isomorphs (Yudovich & Ketris, 2005). The concentration of organic mercury is less than the sulfuric mercury in coals. However, the coals with lower sulfur contents has an increased concentration of organic mercury than the sulfuric mercury (Yudovich & Ketris, 2005).

According to Yudovich & Ketris, (2005), the distribution of mercury in coal is based on the ash yield and also the content of pyritic sulfur. However, increase in the ash values for low ash coals is due to increase in the pyritic sulfur contents masking the determination of the importance of the individual properties. The relationship between the ash yield and the mercury content is based on the genetic form of mercury present in coal. The relationship between the ash yield and terrigenous mercury is linear, but the relation is weak in authigenic coal. There is a strong positive correlation between the contents of mercury and the contents of sulfur in coal concluding that the major form of mercury in coal is pyritic mercury (Cecil et.al., 1979). More than 50 percent of the coals in the United States with increased concentrations of mercury have increased contents of sulfur (Finkelman & Gross, 1999). About 50 percent of the mercury is bound to pyritic sulfur whereas the remaining is bound to the organic matter in coal (Yudovich & Ketris, 2005).

3. CHEMICAL FORM OF MERCURY

Mercury is one of the highly volatile metals that can be volatilized even at very low temperatures of 150 °C. This property attributes to its occurrence in vapor form in the emissions (Galbreath & Zygarlicke, 1996). Less than 2 percent of mercury remains at the bottom ashes after the coal combustion (Meij, 1991).

Mercury is measured as three forms that include elemental/metallic mercury, inorganic mercury and organic mercury. The valence states of mercury emitted from coal fired power plants are elemental, monovalent and divalent mercury. Elemental mercury is highly volatile and evaporates as mercury vapors. According to Selin (2013), elemental mercury gets converted into monovalent and divalent mercury by undergoing series of oxidation reactions and losing electrons. Monovalent and divalent mercury are the oxidative states of inorganic mercury that has the ability to combine with other elements such as chlorine, oxygen or sulfur to form inorganic compounds of mercury that include mercuric sulphide, mercuric oxide, mercuric chloride, etc. (Rischer, 2003). When mercury reacts with carbon, organic mercury compounds such as methylmercury, ethyl mercury, dimethyl mercury, etc. are formed which are the utmost toxic species of mercury that bio-accumulate by a factor up to 105 in the food chain (Gilmour & Henry, 1991).

Elemental mercury is the most stable form that is less likely to be soluble in water leading to a long distance transport (Clarkson, 2002). Among the monovalent and divalent mercury, divalent form is more stable and is more likely to occur in the environment as inorganic forms by the association with chlorine, sulfur and hydroxyl ions (Carpi, 1997). The inorganic compounds also get converted into elemental mercury at temperatures greater than 700 to 800 °C, because they are thermally unstable at the higher temperatures (Otani, Emi, Kanaoka & Matsui, 1984). But when the emissions reach the stack where the temperatures are lower, elemental mercury reacts with other constituents such as oxygen or chlorine present in the emissions to form inorganic compounds (Hall, Lindqvist, & Ljungstroem, 1990).

The type of mercury emitted from the power plants depends upon the total amount of mercury present in the coal used and also the type of control devices such as activated carbon injection or wet lime/limestone flue gas desulfurization equipment used in the plant, the species and the form of mercury that is present in the flue gases (Carpi, 1997). According to Cohen, Artz, & Draxler, (2007), mercury is emitted into the air from coal-fired power plants in three different forms that include elemental mercury (53%), reactive mercury (42%) and particulate mercury (5%). Reactive mercury is the monovalent and divalent mercury that combines with other elements to form inorganic compounds. Particulate mercury is the mercury that is adsorbed on the carbon particles in the flue gas emissions. Less than 5 percent of the mercury emissions contain particulate mercury because most of it is removed by the air pollution control equipment such as fabric filters and electrostatic precipitators (Pacyna & Münch, 1991). It is extremely difficult to control elemental mercury, mercuric chloride and other forms of divalent mercury because they exist in vapor phase and their partial pressures are below the levels required for condensation in combustion flue gas (Prestbo & Bloom, 1995). The percentage of mercury captured in the power plant facilities that do not contain specific mercury control equipment ranges from 0 to 99 percent and the capture rate increases with the increase in carbon content in the exhaust stream (Nebel, 1993).

4. FATE AND TRANSPORT OF MERCURY

According to Stamper, Copeland, Williams, & Spencer (2012), it was estimated that 58,000 pounds of mercury is released in 2010 from coal-fired power plants and the exposure is seen within 300 miles of the power plant. The exposure is greatest at the power plant facility and about 80% of the reactive mercury that travels in the air gets deposited on the land within 1500 miles of the power plant.

The elemental mercury circulates for 1 year in the atmosphere pertaining to its global distribution (Clarkson, 2002). Mercury may also undergo other oxidation and reduction reactions in the atmosphere that leads to the precipitation of mercury (Schroeder, Yarwood, & Niki, 1991). In the stratosphere, the vapor is oxidized to ionic mercury and returns to the earth's surface through rain leading to the distribution of mercury to most of the remote areas in the world (Tchounwou, Ayensu, Ninashvili, & Sutton, 2003). All the forms of mercury eventually gets deposited in water or on land. In the soil, the divalent mercury reacts with the chlorides and hydroxide ions and form inorganic complexes. Increase in the concentration of chlorine decreases the formation of organic mercury in the soil (Olson, Cayless, Ford, & Lester, 1991).

The mercury in the atmosphere and in the soils are washed away into the surface waters through the rainfall. 19 percent of the U.S lakes and rivers were contaminated with mercury during the year 1970 and the low median value was less than 0.5 ppb (Nriagu & Pacyna, 1988). In the water all forms of mercury is converted into more toxic form, methyl mercury by the microorganisms (Stamper, et.al, 2012). Yeasts present in the waters have the capability of the methylation of mercury and also the conversion of ionic form of mercury into elemental form (Yannai, Berdicevsky, & Duek, 1991). Methylation of mercury is also increased by the presence of methyl cobalamin compounds that are synthesized by bacteria (Regnell & Tunlid, 1991). Increase in the concentration of dissolved organic carbon levels decreases the methylation of mercury (Gilmour & Henry, 1991) which is due to the binding of mercury ions to the organic carbon and the decline in the bioavailability of mercury ions for methylation (Miskimmin, Rudd, & Kelly, 1992). According to Björnberg, Lars, & Lundberg, (1988), an increase in the pH of surface waters increases the formation of mercuric sulfide and decrease in the pH increases the formation of methyl mercury. It is due to the fact that acid deposition reduces the activity of sulfide ions.

5. HEALTH EFFECTS OF MERCURY AND ITS COMPOUNDS

All the forms of mercury differ in their degree of toxicity and in their effects on the nervous, digestive and immune systems, and on lungs, kidneys, skin and eyes. The fundamental organs affected by both the acute and chronic effects are the kidneys and the central nervous system (Broussard et.al, 2002). The toxic effects of mercury and its compounds occur by disrupting the tertiary and quaternary structures of cellular proteins. Thus, the effects of mercury are not only seen at organ-system level, but also at the subcellular level (Jaishankar et.al, 2014).

According to EPA, elemental mercury is not classifiable as to human carcinogenicity (Group D), inorganic mercury and methylmercury are possible human carcinogens (Group C) (Mercury compounds, 1992). According to IARC, elemental and inorganic mercury are not classifiable as

to carcinogenicity to humans (Group 3) and methylmercury is considered to be possibly carcinogenic to humans (Group 2B) (Mercury and Mercury Compounds, 1987).

5.1.Acute health effects

Elemental mercury: Acute inhalation of mercury vapor at larger doses causes erosive bronchitis and bronchiolitis that leads to the respiratory system failure and it may also be fatal when acute respiratory distress syndrome develops (Broussard et.al, 2002). It can also cause tremors or erethism by affecting the central nervous system (Garnier et al., 1981). Other central nervous system symptoms are tremors, irritability, insomnia, memory loss, neuromuscular changes, headaches, slowed sensory and motor nerve function and decreased cognitive function (ATSDR, 1999). In the kidneys, the effects range from mild proteinuria to acute renal failure (ATSDR, 1999).

Inorganic mercury: Acute effects are usually seen with ingestion of inorganic mercury. The effects are usually due to the corrosive nature of mercuric salts (Broussard et.al, 2002). According to Barnes et.al, (1980), acute exposure to mercuric salts affects the mucosa of the gastrointestinal system and the kidneys. It causes necrosis of the gastrointestinal mucosa and the symptoms include abdominal pain, vomiting and bloody diarrhea. This can lead to death from septic or hypovolemic shock. The patients who survived develop renal tubular necrosis and the symptoms include anuria. Metallic taste in the mouth, nausea, vomiting and severe abdominal pain are the symptoms associated with acute exposures (ATSDR, 1999).

Organic mercury: Acute inhalation of methyl mercury is very rare and it causes severe CNS effects such as tremors, visual and hearing impairment, paralysis, insomnia and fetal developmental defects (Renzoni, Zino, & Franchi, 1998). It leads to blindness, deafness, speech defects, cerebral palsy and mental retardation in infants (Davis, 2002).

5.2.Chronic health effects

Elemental mercury: The triad of symptoms for chronic elemental mercury exposure are tremors, gingivitis and erethism (Broussard et.al, 2002). Chronic exposure to mercury vapor affects the nervous system and causes weakness and gastrointestinal problems initially and then cause tremors, erethism, behavioral changes, emotional excitability, and loss of memory, insomnia, depression, fatigue, delirium and hallucination (Berglund, Pohl, Olsson, & Bergman, 1988). Oral symptoms such as gingivitis and increased salivary flow can also be present (Nordberg, 2015). Proteinuria occurs when the kidneys are affected (ATSDR, 1999). In children, a syndrome called acrodynia may develop and symptoms include severe leg cramps, irritability, paresthesia, pruritus, diaphoresis, tachycardia, hypertension, photophobia, anorexia, insomnia, poor muscle tone, constipation, diarrhea, painful pink fingers, and peeling of hands, feet and nose (Broussard et.al, 2002).

Inorganic mercury: Chronic exposure to mercuric salts occurs through occupational exposure and the major effect of this exposure is kidney damage (ATSDR, 1999). Effects on the kidneys include renal tubular necrosis and autoimmune glomerulonephritis (Barnes, et.al, 1980). It can also affect the immune system and causes asthma, dermatitis and other hypersensitivity reactions

(de Vos, Abotaga, Liao, Jerschow, & Rosenstreich, 2007). Improper functioning of the thyroid gland may also be seen (Ellingsen et al., 2000). Inhibition of the production of sperms may also occur when the testis are affected (Rao & Sharma, 2001).

Organic mercury: Chronic exposure to methyl mercury damages the CNS. In the central nervous system, the cerebellum, calcarine fissure and the precentral gyrus are usually affected (RAIS, 1998). The early symptoms that are caused by chronic exposure are paresthesia, blurred vision and malaise. Later, it leads to deafness, speech difficulties and constriction of the visual field (ATSDR, 1999). It can also lead to cardiovascular diseases and cancer (Risher, Murray, & Prince, 2002). Exposure of pregnant women to methylmercury gives birth to the infants with CNS effects that range from minor symptoms such as developmental delays and abnormal reflexes to major symptoms such as mental retardation, ataxia, deafness, constriction of the visual field, blindness and cerebral palsy (ATSDR, 1999).

6. TOXICOLOGY

Exposure to elemental mercury: According to Ashe et al., (1953) when rabbits are exposed to 28.8 mg/m^3 of metallic mercury, no death is seen on exposure for 20 hours or less but one in two rabbits died on exposure for 30 hours. Mild to moderate pathologic changes are seen on exposure for 1 to 20 hours and cellular degeneration and necrosis of the lungs is seen after exposure for 30 hours. Exposure of rabbits to 28.8 mg/m^3 mercury vapor for 4 to 30 hours showed marked cellular degeneration and necrosis of the colon and the exposure for 6 to 30 hours showed necrosis of the liver. Exposure of rats to 1 mg/m^3 of metallic mercury vapors for 100 hours per week for 6 weeks, showed congestion in the lungs (Gage, 1961). When the rats are exposed to 3 mg/m^3 of mercury vapor for 3 hours a day, 5 days in a week and for 12 to 42 weeks, there are no significant pathologic changes in the liver and lungs (Kishi, 1978). Autoimmune response in genetically susceptible mice when exposed to mercury vapors for a period of 10 weeks showed the response by general stimulation of the immune system (Warfvinge et al., 1995).

Exposure to inorganic mercury: The range of oral LD50 value in mice is from 25.9 to 77.7 mg/Kg (Kostial et al., 1978). Oral exposure of Long-Evans rats to 2.2 mg/kg/day for a period of three months showed labored breathing, bleeding from nose and difficulty in respiration (Goldman & Blackburn, 1979). Exposure of rats to 28 mg/kg/day for a period of 180 days through drinking water demonstrated an increase in blood pressure and a decrease in cardiac contractility (Carmignani et al., 1992). Dietary exposure to 0.69 mg/kg/day for two years in mice resulted in the ulcers of stomach (Mitsumori et al., 1990). Dietary exposure to 1.1 mg/kg/day for 4 weeks resulted in increased kidney weights in female Wistar rats (Jonker, et.al, 1993).

Exposure to organic mercury: Animal studies on the exposure to methyl mercury are limited. Exposure of monkeys to methyl mercury from birth to seven years of age showed delayed neurotoxicity and at the age of thirteen they displayed abnormal behavior to touch even though their clinical examination showed normal results (Rice, 1996).

7. EXPOSURE TO MERCURY EMISSIONS

7.1.Route of exposure and the amount of absorption

Exposure through inhalation: 97.4 percent of the total elemental mercury absorption occurs through inhalation of the contaminated air near the coal fired power plants (Hursh, Clarkson, Miles, & Goldsmith, 1989). Absorption of inorganic mercury through inhalation route is very low because of the ease of elimination of the particles by the muco-ciliary system of the airway (Friberg & Norberg, 1971). The data is limited for humans but it was estimated that about 40 percent of the absorption of inorganic mercury occurs through inhalation in dogs (Keating, 1997).

Exposure through ingestion: Less than 0.01 percent of the ingested elemental mercury is absorbed from the gastrointestinal tract (Clarkson, 2002). Exposure to the inorganic mercury occurs through the ingestion of contaminated soil or water (ATSDR, 1999). 7 to 15 % of the inorganic mercury is absorbed through intestine after ingestion (Friberg & Norberg, 1971; Keating, 1997). Ingestion of fish and other foods that contain methyl mercury leads to the absorption of about 95 percent of methyl mercury from the gastrointestinal tract (ATSDR, 1999).

Dermal exposure: Exposure of mercury vapors through dermal route is minimal and it accounts for only 2.6 percent of the total elemental mercury absorption (Hursh, Clarkson, Miles, & Goldsmith, 1989). Exposure to the inorganic compounds of mercury occurs through the dermal route while handling contaminated soil and water and only 2-3 percent of the total is absorbed (ATSDR, 1999; Keating, 1997).

7.2.Distribution and excretion of mercury

Elemental mercury: When mercury vapors are inhaled, about 80 percent of the mercury enters the kidneys and brain via blood stream and remains in the body for weeks to months causing acute and chronic health effects (ATSDR, 1999). In the brain, it is converted into inorganic mercury and gets trapped. It can pass through the placenta and affect the developing fetus (Grandjean et al., 1992). The average elimination half-life of elemental mercury is 58 days; about 7 to 14 percent is eliminated within a week of exposure through exhalation and approximately 80 percent is eliminated through urine and feces by conversion into inorganic mercury (Keating, 1997).

Inorganic mercury: 10 to 40 percent of the ingested inorganic mercury accumulates in the kidneys (ATSDR, 1999). It is highly difficult for the inorganic mercury to pass the blood brain barrier and blood placental barrier, but it may reach an infant through the breast milk (ATSDR, 1999). The elimination half-life of inorganic mercury is from 49 to 96 days; about 85 percent of the ingested inorganic mercury is eliminated in the feces within 2 days and the absorbed inorganic mercury is eliminated in weeks to months through urine (Keating, 1997).

Organic mercury: 1 to 10 percent of the absorbed methyl mercury is distributed in the blood. Methyl mercury rapidly crosses the brain and placental barrier. In the brain, it is converted to inorganic mercury and gets trapped leading to the acute and chronic health effects (ATSDR, 1999). The elimination half-life of methyl mercury is 45 to 90 days; about 90 percent is excreted through feces in the form of inorganic mercury (Keating, 1997).

7.3.Biomarkers of exposure

Chronic and low dose exposures to any form of mercury can be determined by collecting the urine samples. Acute and high dose exposures can be measured by collecting the blood samples (Cherian, Hursh, Clarkson, & Allen, 1978). Recent exposures can be detected in both blood and in urine (Naleway et al., 1991). The half-life of mercury in blood is only 3 days that gives an indication for the detection of recent exposures only. In contrast, the excretion in urine lasts from days to months which is a reliable biomarker for the identification of increased levels of mercury (Naleway et al., 1991).

When individual species of mercury are taken into consideration, urinary samples shows the appropriate levels of inorganic and elemental mercury exposure because only a small fraction of organic mercury is excreted through urine (Yoshida, 1985). The average concentration of mercury in blood for general population is 0.002 mg/liter (Nordberg et al., 1992). Exposure to the airborne mercury to a concentration of 1 mg/m³ continuously for a period of 8 hour/day results in the average concentration of 1.4 mg/liter in urine and 0.48 mg/liter in blood (Substances, 2002).

8. MEASURES TO REDUCE THE MERCURY EMISSIONS FROM COAL-FIRED POWERPLANTS

8.1.Activated carbon injection

Injection of dry powdered or wet slurred carbon into the flue gas before the entry of flue gas into the air pollution control equipment increases the particulate carbon in flue gas thereby increasing the adsorption of mercury and its removal by the air pollution control equipment (Carpi, 1997). Mercury adsorption to carbon is also dependent of the exhaust stream temperature (USEPA, 1993). Increase in the temperature of the exhaust stream increases the adsorption of mercury to carbon. But an inverse relationship exists between the bag house temperature which is approximately 120 to 200 °C and the reduction of mercury emissions by the activated carbon injection (Miller et al, 1994). More than 90 percent of the mercury is removed when the activated carbon injection rate is 100 mg/ m³ (Carpi, 1997). The carbon injected power plants showed a reduction of 70 to 90 percent of mercury emissions when compared to the other plants that showed only a reduction of 30 to 65 percent of emissions (Plasynski, Litynski, McIlvried, & Srivastava, 2009). There is a positive correlation between the removal of divalent mercury and the content of carbon in the flue gas (USEPA, 1993). Only a partial amount of elemental mercury is captured by the carbon particles (Miller et al., 1994). According to Felsvang et.al. (1993), the activated carbon particles catalyze the formation of divalent mercury from elemental mercury thereby eliminates the amount of elemental mercury.

8.2.Wet lime/limestone flue gas desulfurization:

This procedure removes about 52 percent of the mercury (Meij, 1991). Injection of limestone for the power plants that do not have wet scrubbers removed up to 56 percent of the mercury emission whereas those that were not injected removed only 18 percent proving that limestone injection is cost-alternative to activated carbon injection (Madden & Holmes, 1998). Due to its high water solubility, divalent mercury can be easily removed from this wet system compared to that of the elemental mercury. According to Vogg, Braun, Metzger, & Schneider, (1986), when the pH is greater than three and the chloride ion concentration is less than 0.1

Moles, the sulfur dioxide present in the flue gas disrupts the reduction and removal of divalent mercury leading to the formation of elemental mercury vapors that escape in the exhaust stream. Thus the capture ability depends upon the pH and chloride ion concentrations.

9. CONCLUSION

The mercury emissions from coal-fired powerplants are increasing in the United States due to the increase in the production of energy. In order to reduce the negative effects by the coal-fired power plants, alternative sources of energy such as renewable sources of energy should be used. According to USEIA (2015), the total number of coal power plants reduced from 629 to 518 whereas the number of renewable sources of energy increased from 741 to 2,299 from 2003 to 2013. The use of renewable sources such as solar, wind, hydrothermal and geothermal energies can replace the total number of coal fired power plants and thus can reduce the estimated 50 percent of the anthropogenic mercury emissions from coal fired powerplants. By using the controlled technology and alternative sources of energy, the risk of this neurotoxic heavy metal can be minimized.

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